

## ***Interactive comment on “Interannual Variability of Ammonia Concentrations over the United States: Sources and Implications” by Luke D. Schiferl et al.***

### **Anonymous Referee #1**

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Schiferl et al. use the GEOS-CHEM model to simulate ammonia concentrations across the U.S. during the period 2008-2012. They evaluate model performance by comparison with in situ (including AMoN) and IASI satellite measurements, use the model to examine factors driving interannual ammonia concentration variability, and infer from model simulations how emissions variability affects PM<sub>2.5</sub> formation and reactive nitrogen deposition. While not entirely novel, the manuscript builds upon earlier model-observation intercomparisons and provides a more comprehensive look especially at factors influencing year-to-year variability in ammonia concentrations. The article is generally well written and clearly describes approach, hypotheses, and findings. There are several items that should be addressed to improve the manuscript:

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1. In the abstract (line 20, p. 1), the authors should be more quantitative in summarizing the model bias in simulating observed ammonia concentrations.
2. p. 2, line 13: the relevant parameter here is sulfate, not SO<sub>x</sub>. An environment can have lots of SO<sub>2</sub> without affecting ammonium nitrate formation, for example if there is insufficient oxidant to promote reaction of SO<sub>2</sub> to sulfate. It is only when that SO<sub>2</sub> is oxidized to sulfate that there is an effect.
3. The authors could make a stronger case for the importance of their work by increasing emphasis in the introduction on the increasingly important contributions ammonia/ammonium are making to reactive N deposition. While U.S. NO<sub>x</sub> reductions are reducing oxidized nitrogen deposition, U.S. ammonium deposition has been increasing. The source of this increase is not well understood. The work presented here could help lay a foundation for better future understanding that change.
4. The paper's focus primarily on summer ammonia is somewhat disappointing. While usable satellite obs are more limited in other seasons, some of the most interesting effects for PM are, as the authors point out, at cooler times of year. The focus on August emissions in Fig. 1 is also a little disconcerting. Major fertilizer emissions from the U.S. heartland (Iowa, Illinois, Indiana, etc...) are missing from this figure because they occur in spring, for example, while this figure just shows August. At a minimum, some additional discussion about how emissions change regionally throughout the year would be helpful to the reader.
5. Considerable attention has been focused in the past couple years on improved treatments of ammonia deposition in models, especially through the incorporation of bidirectional flux schemes. It is surprising that this issue is not mentioned at all in this manuscript, particularly since such "bidi" treatments tend to reduce ammonia deposition, especially near sources, helping models better match observations. At a minimum the authors should discuss the deposition scheme used in their simulations and outline shortcomings of the treatment for ammonia dry deposition.

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6. The column concentrations in Fig. 2 show a surprising depiction of very high values along the coastal region of the Pacific Northwest. What do the authors think of this? This is not a region typically identified with high ammonia concentrations and I note that the number of retrievals here is fairly small. Do the authors think this depiction is realistic?

7. p. 11, line 14: Should the 2.8 ppb mentioned here be the 2.5 ppb mean simulated at AMoN sites?

8. p. 11, line 18: I would be reluctant to directly state that the "AMoN network as a whole has a sampling bias." The goal of the AMoN network is not a statistical sampling to obtain the average U.S. concentration. Please qualify your statement so that someone does not quote you out of context.

9. The 3-site comparison with the model nicely illustrates some of the characteristics of regions where the model predicts ammonia concentrations better or worse. Very nice. I suggest adding plots with similar comparisons for your other sites to supplemental information as many readers might find this useful.

10. p. 13, lines 18-19. It is unclear how you know the LP AMoN site is not showing fire influence. Certainly this site is influenced by upslope transport from the Front Range region for some hours almost every day during the summer.

11. I found myself wondering while reading why you used NEI 2005 rather than NEI 2011 ammonia emissions as your base case. I wondered this even more after seeing in section 4.4 where you compared the two and found NEI 2011 offered some improvements. Please justify.

12. Section 5.1 on effects of SO<sub>2</sub> and NO<sub>x</sub> emissions reductions is very interesting, but you need to discuss whether the model performance for sulfate and nitrate/nitric acid is good enough to reliably interpret the findings to such precision (e.g., change of 32%/0.17 ppb).

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13. The discussion of meteorological variability effects in section 5.2 is also interesting, but could be strengthened by examining the model phase partitioning to support your arguments about the effect of that phenomenon on ammonia concentrations.

14. p. 17, lines 25-27. The sentence beginning "Nearly all changes to the ammonia concentrations..." is confusing since it might make the reader think the volatilization scaling effect is affecting phase partitioning in the atmosphere rather than changes in ammonia volatilization from sources.

15. Please specify that the concentrations in equation 1 are in molar units for clarity.

16. The discussion of the Gas Ratio (GR) on p. 19 needs to be revised. First, the statement that  $GR > 1$  implies little potential for further ammonium nitrate formation is an oversimplification. Yes, this is true for  $GR \gg 1$ , but the dropoff in effect is more gradual than implied since the equilibrium concentration of ammonium nitrate is proportional to the product of the ammonia and nitric acid concentrations. This product can still grow as GR increases above 1 but will start to taper off for much larger values. By choosing  $GR = 1$  as a de facto cutoff for regions/times where ammonium nitrate formation may respond to ammonia concentrations you are oversimplifying what is really a more gradual change. Given how important this issue is to considering future policy re: ammonia emissions, I think you should be more careful in how you describe this effect.

17. While ammonium nitrate formation is limited by warm summertime temperatures, it would be worth mentioning that it might be important overnight as T drops and RH rises and that other ammonium salts (e.g., ammonium oxalate) might be important during this more photochemically active time of year.

18. The authors do a nice job summarizing the needs for better observational constraints on ammonia concentrations in the future. I would add that such constraints should also feature (1) higher time resolution measurements and (2) measurements of both gas and particle phase ammonia/ammonium to (1) provide a better basis for comparison with model simulations with reduced variability in meteorological conditions

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and source impact and (2) to better constrain the total NH<sub>x</sub> budget.

19. The x-axis timelines in Figure 8 should be better labeled/identified.

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